

Final Technical Report:

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Use of airborne radon measurements to recognize long range
transport events in the mid and upper troposphere.

Award Period: 1 February 1988 - 31 May 1989

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Summary

The continuing increase in pollution sources and the resultant increase in the global abundance of pollutants such as sulfur dioxide, non-methane hydrocarbons, and other volatile organic compounds has led to a need to improve our understanding of the global-scale transport of these compounds. Radon, because of its surface source, short (3.8 day) half life, and immunity from removal by chemical or physical scavenging is ideally suited as a tracer for the long-range transport of many pollutants, and when measured from ships has been used for many years as an indicator of the movement of polluted continental air over the oceans. A detailed meteorological analysis, concluded in this study, was made for several air masses sampled aboard the NASA C-141 aircraft in order to establish the source and transport mechanism of radon-rich air observed on several occasions in the upper troposphere over the eastern Pacific Ocean. These analyses, which were incorporated in a manuscript and submitted for publication, showed that the radon-rich air sampled originated in the Asian boundary layer, ascended in cumulus updrafts, and was carried eastward in the fast moving polar jet stream, a mechanism consistent with the rapid long-range transport of many chemically active pollutant species as well.

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Summary

- 1.0 Introduction and background
- 2.0 Preparation and presentation of the manuscript: The China Clipper: Fast advective transport of radon-rich air from the Asian boundary layer to the upper troposphere near California, Mark A. Kritz, Jean-Claude Le Roulley and Edwin F. Danielsen, (Tellus 42B, 46-61, 1990).
- 3.0 References

1.0 Introduction and background.

Radon, because of its surface source, short (3.8 day) half life, and immunity from removal by chemical or physical scavenging is ideally suited as a tracer for the long-range transport of many pollutants, and when measured from ships has been used for many years as an indicator of the movement of polluted continental air over the oceans (e.g. Prospero and Carlson, 1970; Kritz and Rancher, 1980; Polian et al., 1986.)

Atmospheric radon concentrations are highest near the ground, where concentrations in the range 100-300 pCi/SCM (standard cubic meter) are frequently observed (Gold et al., 1964; Cox et al., 1970; Lambert et al. 1982). Concentrations in the free troposphere are typically an order of magnitude lower, decreasing, in the mean, with altitude to values on the order of 1 or 2 pCi/SCM in the upper troposphere (Wexler et al., 1956; Wilkening, 1970; Moore et al., 1973). This decrease of mean concentration with increasing altitude is similar to that which would arise from simple mixing, and for this reason vertical profiles of radon have also been used to estimate the rate of vertical mixing, or transport of boundary layer air into the free troposphere (e.g. Jacobi and Andre, 1963; Liu et al., 1984). In one instance a balloon-borne instrument (Jegou, 1983) was used to make radon measurements in the lower stratosphere, in order to seek evidence (none was found in that particular study) of the movement of tropospheric air into the stratosphere.

In 1982, in cooperation with Jean-Claude Le Roulley of the French Centre des Faibles Radioactivites laboratory, my research group installed a radon measurement instrument aboard the NASA Kuiper Airborne Observatory. The Kuiper Airborne Observatory, or KAO, is a C-141 aircraft which has been modified to carry a large infrared telescope. It is based at Moffett Field and operated by NASA as a national astronomical facility. The KAO makes about 90 flights per year, flying to altitudes as high as 45,000 feet in order to get above the tropospheric dust and water vapor which would interfere with its observations. This platform was almost ideally suited for our study, whose goal was to make radon measurements in the upper troposphere and lower stratosphere at mid-latitudes, in order to seek evidence for the mid-latitude movement of tropospheric air into the stratosphere.

Approximately 50 (piggyback) flights were made in the course of this investigation, over the period 1982-1986. About a third of these were developmental, in which we were conducting systematic background studies or testing instrument performance. The remaining 35 or so flights were scattered over all four seasons of the year, in both the upper troposphere and the lower stratosphere, and over a wide range of geographical locations.

In the course of these flights we never did observe a detectable quantity of radon in the lower stratosphere, and so did not obtain any evidence of the movement of tropospheric air into the stratosphere. We did, however, on several occasions observe unexpectedly high concentrations of radon in the upper *troposphere*---values which were sometimes more than an order of magnitude greater than the mean values of 1-2 pCi/SCM which had been believed to dominate at these levels.

In order to understand this phenomenon, we grouped our flights by season and location. When this was done we found that we had made most of our flights, and most of our upper tropospheric measurements in the summers of 1983 and 1984. Radon concentrations greater than 16 pCi/SCM were observed on 6 of the 13 flights made during these two summertime periods, and in 9 of the 61 individual radon measurements. A frequency distribution plot of these 61 observations showed a distinct bimodal distribution, with approximately 2/5 of the values falling close to 1 pCi/SCM, the expected value, and approximately 3/5 in a high concentration mode centered at about 11 pCi/SCM.

The next step was to explain how this radon-rich air came to be in the upper troposphere. Clearly it originated in the continental boundary layer, since soils are by far the dominant source atmospheric radon (Turekian et al., 1977). In order to elucidate the transport mechanism involved, we joined forces with a nationally pre-eminent meteorologist and meteorological analyst, Dr. Edwin Danielsen of the Space Science Division of the NASA Ames Research Center. We had in fact been working with Dr. Danielsen for some time on related questions of long-range transport and troposphere to stratosphere exchange, but because these KAO observations were so unprecedented, and showed promise of providing dramatic evidence of a rapid, cohesive long-range movement of a boundary layer air mass, we decided to devote a special effort to the understanding these findings. This effort spanned several years, and was covered by several

research projects. The project reported on here focused on the preparation of a manuscript describing these measurements and our meteorological analyses, and submitting it for publication.

2.0 Preparation and presentation of the manuscript:

The China Clipper: Fast advective transport of radon-rich air
from the Asian boundary layer to the upper troposphere near California, by
Mark A. Kritz, Jean-Claude Le Roulley and Edwin F. Danielsen (Tellus 42B, 46-61, 1990).

The meteorological analyses focused on two of the six flights during which exceptionally high radon concentrations were measured. Both of these flights were made in the summer of 1983, in the course of a deployment of the KAO to Oahu, Hawaii: 28 July, 1983; and 31 July, 1983. The 28 July radon measurements were made in the course of a flight to the northwest of Hawaii, while the measurements on the 31 July flight were made while the plane was flying between Hawaii and Moffett Field.

The probable source regions for the air masses sampled during these flights were determined by constructing trajectories from 22 consecutive analyses, made at 12 hour intervals, of the Montgomery stream function, ψ_m , on a constant potential temperature (θ) surface. The sources of the information needed to perform these analyses included satellite photos of the region, synoptic analyses at several levels obtained from the Naval Postgraduate School and the National Weather Service, and vertical temperature soundings obtained from the National Weather Service. These analyses are presented in some detail in the attached publication [Kritz et al., 1990] and are not repeated here.

The trajectory analyses presented in that paper showed that the air sampled in the upper troposphere on the flights of 28 and 31 July with the highest radon concentrations had been located in the upper troposphere over Asia some two to four days prior to sampling. In those analyses it was also seen that the rapid eastward transport occurred in regions where cold cirrus clouds were present in the satellite infrared photographs. The implication of these results is that the radon-rich near surface air was transported upward with the water vapor that produced these clouds.

The most probable for of vertical transport in this situation is within cumulonimbus updrafts originating in the boundary layer. This hypothesis was tested by examining the Nimbus 7 satellite observations for 24 and 29 July, the dates when the trajectories passed over north-eastern China. On both of these dates the satellite photos showed active convective storms in this area, confirmed by surface-level observations reported on the synoptic analysis for the region. Thus, as discussed at greater length in the paper, it is highly probable that the radon rich air observed over the northeastern Pacific on our flights of 28 and 31 July originated in the Asian boundary layer, ascended in cumulus updrafts, and was carried eastward in the fast moving polar jet stream---a mechanism consistent with the rapid long-range transport of many chemically active pollutant species as well.

The final preparation of the manuscript and figures was carried out in the spring and summer of 1988. The manuscript was sent to the journal office on October 28, 1988. Additional revisions were made in the spring of 1989, following the suggestions of the reviewers, and the manuscript was accepted for publication on June 5, 1989.

In addition to presentation in a peer-reviewed paper, these results were also presented at a the September, 1988 Session on Radon and its Daughters in Atmospheric Problems, held in connection with the International Congress of Geochemistry and Cosmochemistry. Our presentation received a great deal of attention from the global modelers attending that meeting, as the systematic occurrence of high radon concentrations in the upper troposphere was not predicted by their models, and so raised the possibility that the transport parameterizations used in those models was incorrect or incomplete. Thus this research grant closed not only with the acceptance for publication of our unexpected findings and the associated meteorological analysis and explanation, but also with the suggestion that these results could have a significant impact on the formulation of the global models used to assess global pollution and climatic change.

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